SUPREME COURT OF THE STATE OF NEW YORK
COUNTY OF STEUBEN

In the Matter of the Application of

SIERRA CLUB, CONCERNED CITIZENS OF ALLEGANY COUNTY, PEOPLE FOR A HEALTHY ENVIRONMENT, INC., JOHN E. CULVER, AND BRIAN AND MARYALICE LITTLE,

Petitioners,

For a Judgment Pursuant to Article 78 of the Civil Practice Law and Rules,

–against–

NEW YORK STATE DEPARTMENT OF ENVIRONMENTAL CONSERVATION, BASIL SEGGOS, COMMISSIONER, AND HAKES C&D DISPOSAL INC.,

Respondents.

AFFIDAVIT OF RAYMOND C. VAUGHAN IN SUPPORT OF THE VERIFIED PETITION

Index No. E2017-1384CV

State of New York,
County of Erie, ss.:

RAYMOND C. VAUGHAN, being duly sworn, deposes and says:

1. I am a Professional Geologist (NY license no. 258) and Environmental Scientist with a Ph.D. in Geology from SUNY Buffalo. I am very familiar with radioactive substances or "radionuclides," sometimes also called "radioisotopes." I am familiar with the physical and quantitative properties of these radionuclides and the alpha, beta, and gamma radiation they emit, including quantitative measures such as activity (expressed in Curies, for example), specific activity, and half-life, and also including relationships such as secular equilibrium that may occur when a parent-progeny relationship exists among different radionuclides. Note that a parent-progeny relationship is sometimes called a "parent-daughter" relationship. Note also that the words "activity" and "radioactivity" can be used interchangeably when referring to a quantitative measurement – as expressed, for example, in picocuries per gram (pCi/g) or

picocuries per liter (pCi/L).

2. My familiarity with such properties, measures, and relationships is based partly on work relating to radionuclides that I performed during the twelve years I was employed as an Environmental Scientist at the NYS Attorney General's Office (2000-2012), partly on consulting work done for the Seneca Nation of Indians to review and interpret radiological test results (2016), and partly on technically-oriented volunteer work I have performed for several decades as a member of the Coalition on West Valley Nuclear Wastes (1978-2006) and also as a member of the West Valley Citizen Task Force (1997-present). I was appointed to the West Valley Citizen Task Force (CTF) by the U.S. Department of Energy and the New York State Energy Research and Development Administration (NYSERDA), and on one occasion in 2006 I testified on behalf of the West Valley CTF before the U.S. Nuclear Regulatory Commission in Rockville, MD. I have also spoken on behalf of the West Valley CTF at meetings such as the Council of State Governments/Blue Ribbon Commission public meeting in Boston (2011) and the National Transportation Stakeholders Forum meeting in Buffalo (2013). My CV is attached as Exhibit A.

3. My familiarity with such properties, measures, and relationships is supported in part by my understanding and frequent use of numerical methods and relationships in various fields of science and technology.

Overview of issues and sources

4. I have reviewed reports/information on both Hakes landfill and the Chemung County landfill. The latter is a reasonable proxy for Hakes and is a useful supplement to the limited information that is available for Hakes. In particular I have reviewed:

(a) the current use of radiation monitoring devices to measure and/or detect the radionuclide known as Radium-226 in truckloads of "fracking" waste

being delivered to landfill disposal sites, particularly the *Truck Monitor Correlation Report*, CoPhysics Corp., June 7, 2015 (hereinafter "Truck Monitor Correlation Report"), attached as Exhibit B, and

(b) recent measurements of Radium-226 and other radionuclides in leachate from Hakes landfill, particularly the series of reports with the PDF document names *51D03_Hakes_Leachate_Radiological…* for second-quarter (2Q) and fourth-quarter (4Q) testing from 2Q 2012 to 2Q 2017, as submitted to NYSDEC by either Casella Waste Services or On-Site Technical Services, Inc. (hereinafter "Hakes Leachate Test Reports"), attached as Exhibits C-M, and

(c) recent measurements of Radium-226 and other radionuclides in leachate from the Chemung County landfill, particularly *Chemung County Landfill, Elmira, New York – First Quarter 2017 Leachate Radiological Test Results*, On-Site Technical Services, Inc., submitted to NYSDEC April 5, 2017 (hereinafter "Chemung Leachate Test Report"), attached as Exhibit N.

5. Based on my review I conclude that:

a) The current use of radiation monitoring devices to characterize truckloads of "fracking" waste entering landfill disposal sites is not providing a reliable measure of Radium-226. Secular equilibrium between Radium-226 and its progeny (particularly the radionuclides Lead-214 and Bismuth-214) is a crucial factor whose importance has not been sufficiently recognized. This has resulted in substantial uncertainty about the concentrations and quantities of Radium-226 entering landfills such as Hakes and Chemung County.

b) Measurements of leachate from Hakes and Chemung County

landfills are not providing a reliable measure of Radium-226 and/or Radon-222. The leachate test reports and their interpretations of the data have failed to assess and determine why Lead-214 and Bismuth-214 test results are orders of magnitude higher than the results for Radium-226.

6. For the current practice of using radiation monitoring devices to characterize truckloads of "fracking" waste, there is no evidence that full secular equilibrium, or a consistent degree of secular equilibrium or disequilibrium, exists between Radium-226 and its progeny such as Lead-214 and Bismuth-214. On the contrary, secular equilibrium is disrupted by escape of the gas-phase radionuclide known as Radon-222, with the loss of radon being highly variable and inconsistent. The effects of this are not acknowledged or addressed. Thus, the Truck Monitor Correlation Report does not provide either a reliable limit or an accurate measure of Radium-226 entering the landfill.

7. For the current practice of measuring radionuclide concentrations in leachate from Hakes and Chemung County landfills, there are two alternative explanations for the mismatch between reported Lead-214 and Bismuth-214 test results and reported Radium-226 test results. These involve either underreported levels of Radium-226 or high, unreported levels of Radon-222 in the landfill leachate. The latter is more likely, but additional testing is needed to confirm this. Informed decisions could then be made about addressing the high levels of Radon-222 in the leachate and seeking a better understanding of the landfill's inventory of Radium-226 from which the Radon-222 is generated.

Scientific background: Relevant radiological principles

8. A good summary of the relevant science can be found in the following expert explanation which was posted online in response to an online inquiry that was related to, but not

identical to, the points at issue here:

> Most of the significant gamma radiation from 226Ra decay comes from the radioactive progeny 214Pb and its daughter, 214Bi. These are produced following the decay of 226Ra to 222Rn, which then decays to 214Pb. Since 222Rn is a gas it will escape, to varying degrees, from unsealed sources, and the gamma radiation from the 214Pb and 214Bi may not be significant in such cases. In sealed sources that prevent leakage of 222Rn, the 222Rn, 214Pb, and 214Bi each reach the same activity level as that of the 226Ra within a few weeks of preparation of the source….
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> (Answer posted 27 September 2005 by George Chabot, PhD, CHP, providing an expert online answer to the question, "What value should be taken as the average gamma energy of 226Ra?" at the Health Physics Society website [https://hps.org/publicinformation/ate/q4817.html]. The web page is attached hereto as Exhibit O.)

Note that the above-quoted explanation by Chabot refers to Radium-226 as 226Ra, Radon-222 as 222Rn, Lead-214 as 214Pb, and Bismuth-214 as 214Bi. He also uses the term "daughter" rather than "progeny."

9. The relevant science and Chabot's explanation are rooted in the fact that Radium-226, Radon-222, Lead-214, and Bismuth-214 are all members of the Uranium-238 decay chain. See Exhibit P, which shows that Uranium-238 decays to Thorium-234, and so on. For the present purpose, the decay chain from Radium-226 onward is most relevant. Radium-226 decays to Radon-222, which in turn decays to Polonium-218, which in turn decays to Lead-214, which in turn decays to Bismuth-214, and so on. Given this relationship, Radium-226 can be called the parent, and Radon-222, Polonium-218, Lead-214, and Bismuth-214 can be called the progeny of Radium-226. In effect, there is a "pipeline" from Radium-226 that delivers the first two generations of progeny (Radon-222 and Polonium-218), from which Lead-214 and Bismuth-214 will in turn be created as the third and fourth generations. However, the "pipeline" can be considered leaky if Radon-222 escapes as a gas, in which case Lead-214 and Bismuth-214 will be created at some downwind location rather than in close proximity to the parent Radium-226. As

stated above by Chabot, "Since 222Rn is a gas it will escape, to varying degrees, from unsealed sources, and the gamma radiation from the 214Pb and 214Bi may not be significant in such cases," meaning that the gamma radiation from Lead-214 and Bismuth-214 would not contribute significantly in such cases to a gamma radiation measurement of the parent Radium-226. This is important for the reason stated in Chabot's first sentence: "Most of the significant gamma radiation from 226Ra decay comes from the radioactive progeny 214Pb and its daughter, 214Bi."

10. Thus, in measuring gamma radiation from Radium-226, it's crucial to know whether Lead-214 and Bismuth-214 are present in the sample along with Radium-226. However, even if they're absent due to a "leaky pipeline," a sample collected for Radium-226 analysis can be put in a sealed container, thereby allowing "ingrowth" of the progeny. Keeping a sample in a sealed container for about 21 days or more is sufficient to allow ingrowth and thereby to reestablish "secular equilibrium" for the first four generations of progeny, including Radon-222, Polonium-218, Lead-214, and Bismuth-214. As stated by Chabot, "In sealed sources that prevent leakage of 222Rn, the 222Rn, 214Pb, and 214Bi each reach the same activity level as that of the 226Ra within a few weeks of preparation of the source."

11. This statement by Chabot is illustrated graphically by slide 12 of the Oak Ridge Institute for Science and Education (ORISE) presentation, *Radiological and Chemical Properties of Uranium*, available online from the U.S. Nuclear Regulatory Commission website (www.nrc.gov/docs/ML1122/ML11227A233.pdf), hereinafter "ORISE Presentation," attached hereto as Exhibit Q.

12. An important aspect of the relevant science is the "half-life" of each radionuclide in the decay chain, as shown in Exhibit P. Compared to its progeny, Radium-226 has a long half-life (1600 years). This is much longer than the half-lives of its progeny, including the first four

generations of progeny that are most relevant here: Radon-222 (3.82 days), Polonium-218 (3.1 minutes), Lead-214 (27 minutes), and Bismuth-214 (20 minutes).

13. For the above reasons, Radium-226 in an unsealed container (such as either an uncapped sample vial or the bed of a waste-hauling truck) may be accompanied by relatively little Lead-214 and relatively little Bismuth-214 due to escape of Radon-222 from the container. The loss of Radon-222 will have interrupted the decay chain or the "pipeline" that generates Lead-214 and Bismuth-214. Under these circumstances, the activity or radioactivity of Lead-214 and Bismith-214 in the unsealed container (measured as pCi/g or pCi/L, for example) will tend to be less than the activity of Radium-226 in the container.

14. For the above reasons, Radium-226 which has remained in a sealed container for about 21 days or more will be in secular equilibrium with the first few generations of its progeny, including Lead-214 and Bismuth-214. This means that the activity of Lead-214 and Bismith-214 in the sealed container (measured as pCi/g or pCi/L, for example) will be approximately the same as the activity of Radium-226 in the container. Having Lead-214 and Bismith-214 present will substantially increase the gamma activity as compared to the gamma activity of Radium-226 alone, as discussed below in more detail. As noted by Chabot, "Most of the significant gamma radiation from 226Ra decay comes from the radioactive progeny 214Pb and its daughter, 214Bi."

15. For the above reasons, the activity or radioactivity of Radium-226 which has remained in a sealed container for about 21 days or more cannot be less than the activity of Lead-214 and Bismith-214 in the container. In effect, the "downstream end of the pipeline" cannot generate Lead-214 and Bismuth-214 at a greater rate than is sustained at the "upstream end" of this imaginary pipeline by the decay of Radium-226.

16. For the above reasons, a measurement that shows essentially equal activities for

Lead-214, Bismuth-214, and Radium-226 in a container that has been sealed for about 21 days or more *cannot show whether Lead-214 and Bismuth-214 were present or essentially absent* at the time when the container was sealed. As described on slide 11 of the ORISE Presentation, "Starting with nothing but the parent, the time to reach secular equilibrium is roughly five to ten half-lives of the daughter." In other words, starting with no radionuclides other than Radium-226 in the container when it was initially sealed, ingrowth of progeny will occur as a result of Radium-226 decay. Secular equilibrium with its progeny Radon-222 will be achieved within about 21 days in the sealed container, and secular equilibrium with the next three generations of progeny will likewise be achieved within the same time period. But it cannot be readily determined, based on Lead-214, Bismuth-214, and Radium-226 activity measurements at the end of 21 days or more, whether Lead-214 and Bismuth-214 were essentially absent or already present at some concentration (ranging up to secular-equilibrium concentration) when the container was initially sealed.

17. The general truths expressed above can be applied to the Truck Monitor Correlation Report, Hakes Leachate Test Reports, and Chemung Leachate Test Report.

Unreliability of monitoring trucks for radium concentration at landfill gate

18. The Truck Monitor Correlation Report purports to establish a numerical correlation (i.e., conversion factor) between the concentration of Radium-226 entering a landfill in a waste truckload and the radiation monitor reading at the landfill gate:

> Gamma radiation detectors are routinely used at landfill weighing scales to determine if entering trucks contain unauthorized radioactive materials. However, readings on the monitors cannot easily be related to the concentration of radioactive materials in loads.... Therefore, an actual in-field correlation test was performed to more accurately relate gamma count rate to radionuclide concentration in a load.... A composite sample of the sludge cake was collected from 4 spots near the center of the load (approximately where the detectors were positioned) and was sent to Pace

> Laboratories (NELAP-certified) for gamma spectroscopic analysis after 21-day radon progeny ingrowth.....
>
> (Truck Monitor Correlation Report at 2)
>
> The result of this test of a 30-yard roll-off, filled to near capacity, resulted in a gamma count rate to radium concentration conversion factor or 0.306 KCPS/(pCi/g) over background. For a monitor with a background of 3.6 KCPS (the background occurring during the most recent calibration of the Chemung County Landfill monitor), the count rale corresponding to a 25 pCi/g radium-226 investigation level would be (0.306 x 25) + 3.6 = 11.25 KCPS. Presently the Chemung County monitor's alarm levels arc 10 KCPS sum alarm (sum of both detectors) and a sigma alarm of 110 which equates to approximately 7 KCPS depending on truck speed entering the detection area.... These alarm settings are well within the 11.25 KCPS level corresponding to 25 pCi/g of radium. Therefore, the present alarm settings at the Chemung County Landfill are sufficient to detect a roll-off containing 25 pCi/g or more of radium-226.
>
> (Id. at 5, where KCPS stands for kilocounts per second.)

19. This purported correlation and conversion factor are unreliable because the concentration of relatively strong gamma emitters (such as Lead-214 and Bismuth-214) in the truckload of waste entering the landfill is highly variable and very uncertain for the reasons described above, and because Radium-226 emits only weak gamma radiation in addition to the alpha particles that it emits. As described in an International Atomic Energy Agency report:

> The determination of 226Ra in environmental solids by gamma spectrometry has long been based on the detection of emissions of the radon progeny (222Rn) nuclides, i.e. 214Pb and 214Bi after an ingrowth period of at least 20 days, during which the sample has been hermetically sealed to ensure secular equilibrium between 226Ra and its progeny....
>
> (International Atomic Energy Agency, "Analytical Methodology for the Determination of Radium Isotopes in Environmental Samples," IAEA/AQ/19 (2010) [http://www-pub.iaea.org/MTCD/Publications/PDF/IAEA-AQ-19_web.pdf]).

Since a waste-hauling truck isn't a container that's been sealed for at least 20 or 21 days but may nevertheless contain some quantity of progeny derived from Radon-222 that didn't escape from

the truckload of waste, the use of a gamma monitor to infer Radium-226 concentration in the incoming waste has the potential to be wildly inaccurate and cannot be considered reliable.

20. A relatively tightly packed and/or covered truckload of waste, probably including the sludge cake used for the Truck Monitor Correlation Report, will retain higher concentrations of Radon-222 and other progeny than would be found in a less tightly packed and/or uncovered truckload. See, for example, the description of such ingrowth in *Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) Study Repor*t, Rev. 1, 2016, prepared for Pennsylvania Department of Environmental Protection by Perma-Fix Environmental Services, Inc. (hereinafter "Pennsylvania TENORM Report"), § 5.3. The gamma radioactivity of a relatively tightly packed and/or covered truckload would therefore be relatively high compared to the gamma radioactivity of a loosely packed and/or uncovered truckload carrying the same concentration of Radium-226. If the sludge cake used for the Truck Monitor Correlation Report happened to be relatively tightly packed and/or covered, and thus relatively highly radioactive compared to most other truckloads entering the landfill with the same concentration of Radium-226, this would mean that the report's correlation (indicating that 11.25 kilocounts per second corresponds to 25 pCi/g Radium-226) is unrepresentative and unprotective, allowing truckloads with much more than 25 pCi/g Radium-226 to enter the landfill without exceeding the 11.25 kilocounts per second investigation level. Since the report neither acknowledges this correlation issue nor describes any steps taken to control for the issue, it is difficult to judge from the reported information how high the Radium-226 concentration could be without exceeding the 11.25 kilocounts per second investigation level. In any case, the correlation procedure described in the Truck Monitor Correlation Report is unsupported and cannot be considered reliable based on the information provided in the report.

21. DEC's September 18, 2015 Program Policy Memorandum on "Recommended Permit Modifications and Operating Procedures for Landfills relating to Wastes from Drilling in the Marcellus Shale Formation," attached as Exhibit R, suffers from the same defect of not recognizing the presence/absence/variability of the Lead-214 and Bismuth-214 progeny, and the resulting variability of gamma radioactivity measurable at the landfill gate, in truckloads carrying identical Radium-226 concentrations. See especially §§ 2(b) and 4(b)(v) of DEC's memorandum, which seek to establish or verify a correlation without acknowledging how the Lead-214 and Bismuth-214 progeny affect the intended correlation and without describing any steps needed to control for this issue. DEC's reliance on such a correlation is therefore unfounded.

22. Depending on how much Lead-214 and Bismuth-214 are present in a truckload of Radium-226-bearing waste, the gamma radiation dose measured outside the truck may vary *by almost two orders of magnitude*. In other words, the gamma radioactivity measured outside a truck carrying Lead-214 and Bismuth-214 in secular equilibrium with Radium-226 would be about 60 times greater, or almost two orders of magnitude greater, than the gamma radioactivity measured outside an otherwise equivalent truck carrying no Lead-214 and Bismuth-214. These are the two limiting cases, both of which would show the same radionuclide analysis results for samples collected from within the truckload of waste. The Radium-226 analysis results would be the same because this radionuclide is assumed to be present at the same concentration in both cases. The Lead-214 and Bismuth-214 results would be the same in both cases because of ingrowth and achievement of secular equilibrium during the required sample holding period (at least 21 days). Thus, the initial presence or absence of Lead-214 and Bismuth-214 has no effect on the truckload sample analysis but would have a roughly 60-fold effect on gamma radioactivity

measured outside the truck at the landfill gate. This renders any "correlation" meaningless unless the monitoring procedure at the landfill gate can quantify, and control for, the concentrations of Lead-214 and Bismuth-214 in the load of waste at the time the truck enters the landfill gate.

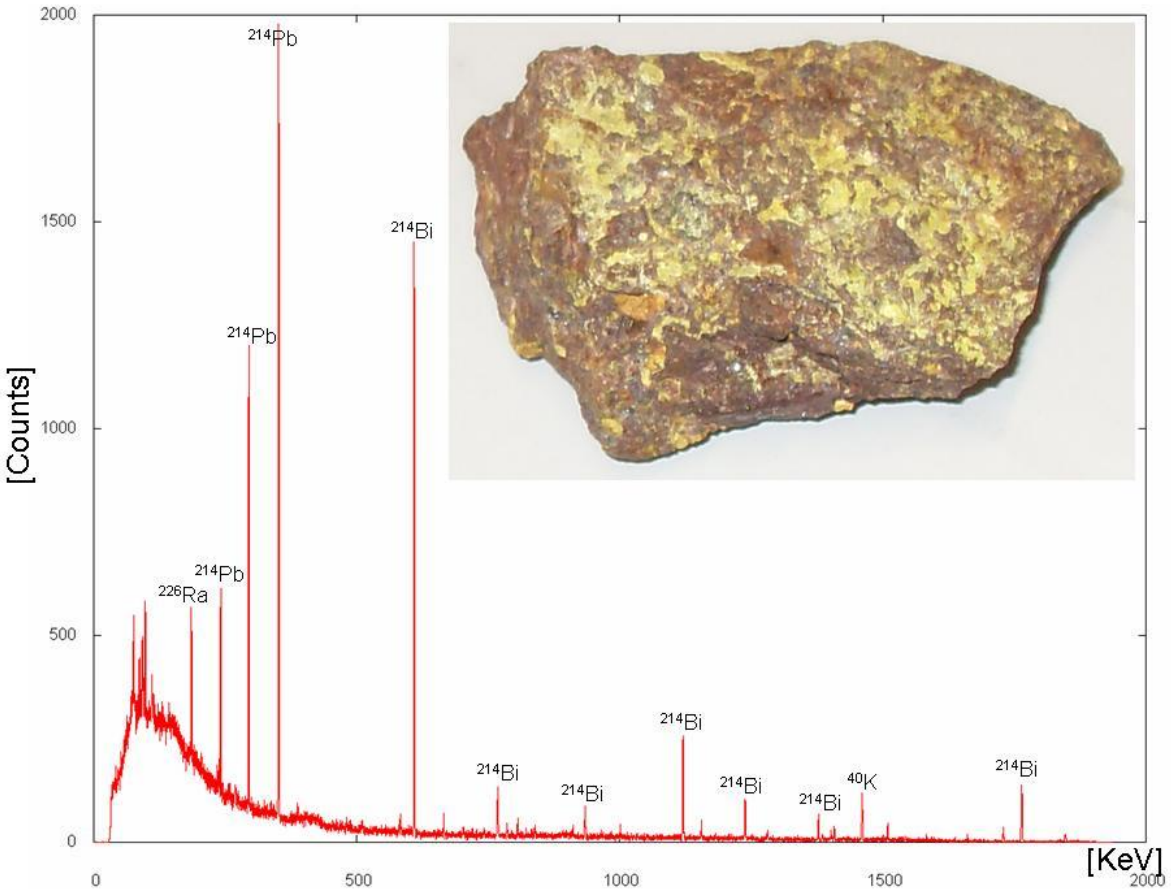
23. The 60-fold variation in the gamma dose measured outside a truck carrying a given concentration of Radium-226 is a combination of two different factors. One factor involves the different gamma yields associated with the three radionuclides; the other is a result of the Lead-214 and Bismuth-214 gamma emissions having greater energy and greater penetrating power than the Radium-226 gamma emissions.

24. The first factor (different gamma yields associated with the three radionuclides) is described in general terms as follows:

> Characteristic gamma rays are emitted in the decay of most radioisotopes; alpha, beta and positron decay and electron capture generally leave the product nucleus in an excited state, which subsequently decays to the ground state with the emission of one or more photons. These gamma rays vary widely in energy and abundance from one isotope to another.
>
> (D. Harris and J. Epstein, eds., *Properties of Selected Radioisotopes*, NASA report SP-7031 (1968), p. 5.)

The "abundance" of gamma rays emitted in the decay of radionuclides can also be called the "gamma yield" of the decay process.

25. The gamma-ray energies and gamma yields associated with Radium-226, Lead-214, and Bismuth-214 decay are available from sources such as D. Delacroix et al., *Radionuclide and Radiation Protection Handbook 2002*; V. Chisté, M.M. Bé, and C. Dulieu, *Evaluation of decay data of radium-226 and its daughters*, International Conference on Nuclear Data for Science and Technology 2007; an online table posted at https://www.cpp.edu/~pbsiegel/bio431/genergies.html; and a graphical representation from https://en.wikipedia.org/wiki/File:Gammaspektrum_Uranerz.jpg which is shown below:



26. As shown in these various sources, decay of Radium-226 is associated with the emission of a relatively weak gamma ray (186 keV), and the gamma yield of this decay process is low, with only about 3.5% of the Radium-226 decay events producing a gamma ray. In other words, each Radium-226 decay emits an alpha particle as the Radium-226 parent atom is transformed into the progeny Radon-222 atom, but gamma rays are emitted in only 3.5% of these transformations or decay events. In other words, there is about a 3.5% probability that a 186 keV gamma ray will be emitted when Radium-226 decays. The 186 keV gamma ray is the only gamma associated with Radium-226 decay. In contrast to this, the decays of Lead-214 and Bismuth-214 may be accompanied by any one of several different gamma energies and yields. Each decay of Lead-214, for example, may be accompanied by emission of a 242 keV gamma ray

(with a gamma yield of about 7%), or by emission of a 295 keV gamma ray (with a gamma yield of about 19%), or by emission of a 352 keV gamma ray (with a gamma yield of about 36%). Each decay of Bismuth-214, for example, may be accompanied by emission of a 609 keV gamma ray (with a gamma yield of about 46%), or by emission of a 768 keV gamma ray (with a gamma yield of about 5%), or by emission of a 934 keV gamma ray (with a gamma yield of about 3%), or by emission of a 1120 keV gamma ray (with a gamma yield of about 16%), or by emission of a 1238 keV gamma ray (with a gamma yield of about 6%), or by emission of a 1378 keV gamma ray (with a gamma yield of about 4%), or by emission of a 1764 keV gamma ray (with a gamma yield of about 16%).

27. It is evident from the foregoing values that the various gamma rays from Lead-214 and Bismuth-214 decay have higher energies than the 186 keV gamma rays from Radium-226 decay. If all three radionuclides are in secular equilibrium, it is also evident that the various gamma rays emitted by Lead-214 and Bismuth-214 will greatly outnumber those emitted by Radium-226, i.e., will be emitted at a substantially higher rate than those emitted by Radium-226. If a monitoring instrument at a landfill gate is counting gamma rays emitted from a truckload of waste (as expressed in kilocounts per second or KCPS), most of the gamma counts will be from Lead-214 and Bismuth-214 rather than Radium-226 if all three radionuclides are in secular equilibrium. Alternatively, if a truckload of waste contains the same concentration of Radium-226 but no Lead-214 or Bismuth-214, the gamma counts will be much lower. The ratio of the gamma counts for these two limiting cases is approximately 40, as can be calculated from the foregoing values. In other words, the gamma count from Radium-226, Lead-214, and Bismuth-214 in a truckload of waste in which all three radionuclides are at secular equilibrium will be roughly 40 times higher than the gamma count from an otherwise equivalent truckload that

contains no Lead-214 or Bismuth-214. This factor of 40 assumes no shielding from the steel walls of the truck. As described below, the shielding of the truck walls will create an even greater difference between these two limiting cases where the Radium-226 concentration remains the same while Lead-214 and Bismuth-214 are either absent, or present at secular equilibrium concentrations.

28. Shielding values for Radium-226, Lead-214, and Bismuth-214 gamma emissions can be calculated from tables of mass attenuation coefficients for iron/steel such as those published by NIST or in the Radiological Health Handbook. A 5-mm thickness of steel, which is an approximate average thickness over all surfaces of a 30-yard roll-off, will attenuate the 186-keV gamma radiation from Radium-226 decay to about half of its unshielded intensity or count rate. The same thickness of steel will attenuate the more energetic gamma radiation from Lead-214 and Bismith-214 decay to about two-thirds of its unshielded intensity or count rate.[1] When these shielding values are combined with the factor of 40 described above, they indicate that the gamma count from Radium-226, Lead-214, and Bismuth-214 in a truckload of waste in which all three radionuclides are at secular equilibrium will be roughly 60 times higher than the gamma count from an otherwise equivalent truckload that contains no Lead-214 or Bismuth-214.

29. Thus, depending on how much Lead-214 and Bismuth-214 are present in a truckload of Radium-226-bearing waste, the gamma radiation dose measured outside the truck may vary by a factor of about 60. This defeats the purpose of using a gamma monitor at the landfill gate and relying on a simple kilocounts-per-second (KCPS) limit. A reliable correlation to a truckload's concentration of Radium-226 cannot be developed unless and until concentrations of Lead-214 and Bismuth-214 are measurable at the moment of entry into the landfill gate. Using

[1] Attenuation depends partly on load-specific information such as distribution of radionuclides within a waste load – which, where known, would allow more precise calculation that incorporates buildup, etc.

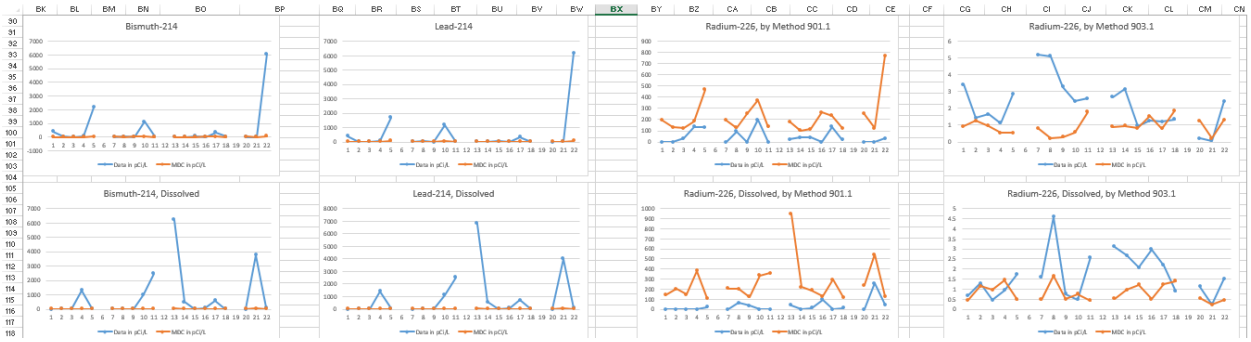
a radioisotope identification device at the landfill gate may be a useful first step toward addressing this problem and developing a reliable correlation, but such a correlation needs to be developed in a transparent, deliberative, and defensible manner, preferably in the context of an EIS process.

Unresolved problems with leachate test results

30. There are significant unresolved problems with the Hakes and Chemung Landfill leachate results reported in the Hakes Leachate Test Reports and Chemung Leachate Test Report. There are at least two problems with these reported results, including (a) high recent radionuclide concentrations and (b) the unrecognized/uninterpreted mismatch between reported Lead-214 and Bismuth-214 test results and reported Radium-226 test results. See results from the Hakes Leachate Test Reports and Chemung Leachate Test Report plotted below (and in Exhibits S-Z), where the horizontal axis on each graph is time, and the graphs show four different time trends.

For Hakes Landfill:

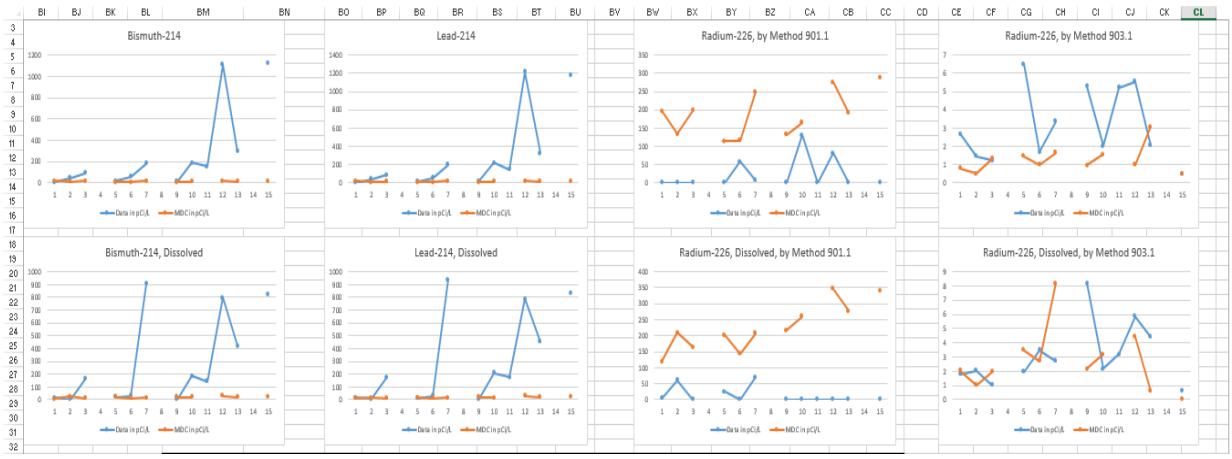
- 1-5 are the 2015-2017 time trend for Cell 3 Leachate
- 7-11 are the 2015-2017 time trend for Cell 4 Leachate
- 13-18 are the 2014-2017 time trend for Cell 5 Leachate
- 20-22 are the 2016-2017 time trend for Cell 8B Leachate



In the above graphs, the blue lines show time trends for the reported test results, while the orange lines show the detection limit (MDC). See also Exhibits S-V for these graphs in larger format.

For Chemung County Landfill:

- 1-3 are the 2015-2017 time trend for Leachate Pond (Combined Leachate)
- 5-7 are the 2015-2017 time trend for Cells I through III Primary Leachate
- 9-13 are the 2015-2017 time trend for Cell IV Primary Leachate
- 15 is the single data point for the 2017 measurement of Cell V Primary Leachate.



In the above graphs, the blue lines show time trends for the reported test results, while the orange lines show the detection limit (MDC). See also Exhibits W-Z for these graphs in larger format.

31. Despite some variability, the overall trend of high radionuclide concentrations in recent leachate test results is evident in the above graphs.

32. The above graphs also show the mismatch between reported Lead-214 and Bismuth-214 test results and reported Radium-226 test results in leachate from both landfills. The results for Lead-214 and Bismuth-214 are robust and mutually consistent, with recent results ranging up to about 1000 pCi/L for Chemung leachate and about 6000 pCi/L in Hakes leachate. See also the much lower activity reported for Radium-226, consistently less than 10 pCi/L when measured by EPA Method 903.1. Whether these Radium-226 test results are reliable is uncertain and cannot be determined unless/until additional testing is done.

33. The question that needs to be resolved is whether Radium-226 in both landfills' leachate is substantially underreported or whether Radon-222 in both landfills' leachate is high and not recognized as such.[2] One or the other of these possibilities (or some combination of the two possibilities) must be true, given the high activities reported for Lead-214 and Bismuth-214 in both landfills' leachate. Stated another way, either the actual Radium-226 activities have ranged up to ~1000 pCi/L in Chemung leachate and up to ~6000 pCi/L in Hakes leachate, or the actual Radon-222 activities have ranged up to ~45,000 pCi/L in Chemung leachate and up to ~270,000 pCi/L in Hakes leachate. Either of these possibilities – or a combination of the two – warrants followup investigation in order to assess impacts to the environment and public health and safety.

34. Details of these two alternatives are reviewed below along with two other possibilities that can quickly be ruled out. These are the only four possible explanations for how the measured activities of Lead-214 and Bismuth-214 could greatly exceed the measured activity of Radium-226 in samples that have remained sealed for 21 days or more:

- The test measurement of Radium-226 activity is erroneous; the actual activity of Radium-226 in the tested samples was much higher than reported, and in reality was approximately the same as the measured activities of Lead-214 and Bismuth-214. Similar activities for all three of these radionuclides would be expected if all three were in secular equilibrium in the sample. Such an error in the measurement of Radium-226 activity is possible but unlikely based on carrier and tracer yields discussed in the accompanying affidavit submitted by D. May. Details of this alternative are discussed below.
- The test measurements of Lead-214 and Bismuth-214 activities are erroneous; the actual activities of these radionuclides in the tested samples were much lower than reported, and in reality were approximately the same as the measured activity of Radium-226. Similar activities for all three radionuclides would be expected if all

[2] Hakes and Chemung leachate testing has generally not included tests for Radon-222.

three were in secular equilibrium in the sample. However, such an error in the measurement of Lead-214 and Bismuth-214 activities is highly implausible and can be ruled out here because the measured Lead-214 and Bismuth-214 activities are robust (well above the detection limit) and mutually consistent.

- Lead-214 and Bismuth-214 activities in the sample are much higher than Radium-226 and Radon-222 activities in the sample. In this circumstance, neither Radium-226 nor Radon-222 would be in secular equilibrium with Lead-214 and Bismuth-214, and the decay rate of Radon-222 in the sample would have been much too low to provide Lead-214 and Bismuth-214 ingrowth at the rate needed to maintain their reported activities. In the absence or near-absence of Lead-214 and Bismuth-214 ingrowth, both of these radionuclides must have been present at extremely high levels at the time of sample collection, and the activities of both radionuclides in the sealed sample must have been constantly declining in accordance with their half-lives – but this can be ruled out based on their relatively short half-lives (~20 minutes). Given the number of half-lives that elapsed during the 21-day period that the samples remained sealed before testing, the activities of Lead-214 and Bismuth-214 in leachate at the time of sample collection would have been impossibly high (hundreds of orders of magnitude higher than their reported test results which ranged up to ~6000 pCi/L), thus allowing this possibility to be ruled out.
- Lead-214, Bismuth-214, and Radon-222 activities in the sample are much higher than the Radium-226 activity in the sample, with Radon-222 being at or near secular equilibrium with Lead-214 and Bismuth-214. Decay of Radon-222 within the sample would provide Lead-214 and Bismuth-214 ingrowth at the rate needed to produce their reported activities at the end of the 21-day sample hold period, but the sample would contain far too little Radium-226 to provide Radon-222 ingrowth at the rate needed for the known ingrowth of Lead-214 and Bismuth-214. Given the near-absence of Radon-222 ingrowth, Radon-222 must have been present at a relatively high level at the time of sample collection, and its activity in the sealed sample must have been constantly and exponentially declining in accordance with its 3.82-day half-life. This is possible; it can't be ruled out based on available

information. Details are discussed below.

35. Thus, there are two alternatives. One is that the actual activity of Radium-226 in the tested samples was much higher than reported due to error(s) in the collection, handling, and/or testing of the samples collected for Radium-226 analysis. The other is that Radon-222 activity (not routinely tested) was high, but Radium-226 activity was not. Distinguishing between these two alternatives could easily be done if samples remained sealed after the original test and could be retested at least several days later. Retesting that showed Lead-214 and Bismith-214 activities similar to the original test results for these two radionuclides would indicate that the first alternative is true or predominant, the reason being that the nearly constant activity of Radium-226, with its 1600-year half-life, would sustain nearly constant rates of Radon-222, Lead-214, and Bismuth-214 regeneration (ingrowth) and decay (activity). Retesting that showed Lead-214 and Bismith-214 activities substantially lower than the original test results for these two radionuclides would indicate that the second alternative is true or predominant, the reason being that the gradually declining activity of Radon-222, with its 3.82-day half-life, would sustain gradually declining rates of Lead-214 and Bismuth-214 regeneration (ingrowth) and decay (activity).

Details of one alternative: Underreported radium concentration in sampled leachate

36. In this alternative, the actual activity of Radium-226 in the tested samples must have been much higher than reported due to error(s) in the collection, handling, and/or testing of the samples collected for Radium-226 analysis. How could such errors arise? Part of the problem may be that an EPA test method for drinking water (Method 903.1) is being used for testing leachate. Leachate samples are aqueous but far less pure than drinking water; they typically contain various dissolved and suspended solids which tend to interfere with sample

preparation. See especially A.W. Nelson, D. May, A.W. Knight, E.S. Eitrheim, M. Mehrhoff, R. Shannon, R. Litman, and M.K. Schultz, "Matrix Complications in the Determination of Radium Levels in Hydraulic Fracturing Flowback Water from Marcellus Shale," *Environ. Sci. Technol. Lett.* **1**, 204−208 (2014). While these authors found radon emanation methods of measuring Radium-226 to be more accurate than wet chemistry methods, the accuracy of an emanation method such as Method 903.1 may be highly dependent on whether Radium-226 will be lost or unrepresentatively partitioned during sample preparation prior to testing. Sample preparation specified in § 8.0 of Method 903.1 includes barium sulfate coprecipitation, which is one of the methods that Nelson et al. found problematic (id. at 206).

37. One possible source of error involves filtration procedures. These cannot be understood in detail from the available information. For example, page 6 of the Chemung Leachate Test Report is a collection record for the C[ell] 5 Primary Leach[ate]; it shows leachate sample collection in a 5-gallon bucket on January 11, 2017. The bottom of the same sheet shows the number of sample containers as 10, evidently indicating that a single bucketful of leachate was poured into ten sample containers, all of which should have contained essentially identical leachate samples. The first of two Chain of Custody forms (id. at 12) shows these ten sample containers being transferred to the ALS Rochester lab along with three other sets of ten from other leachate sample collection points, for a total of 40 containers in all. An instruction printed on the Chain of Custody form says: "Note: Dissolved analysis requires lab filtering."

38. Filtration was evidently performed on half of the 40 containers of samples, and was apparently performed after the ALS Rochester lab transferred the samples to Pace Analytical on January 13, 2017, although no details or specific reference to the filtering process are provided in the report. The second of two Chain of Custody forms (id. at 46-47) shows eight sets of 5 samples

each being transferred to Pace Analytical, half of them marked "dissolved," for a total of 40 containers in all. While the word "dissolved" would tend to indicate that filtration had already been done before ALS Rochester transferred the samples to Pace, this was evidently not the case, as the second Chain of Custody form includes a "Test Comment" that says "Sample Requires In-Lab Filtering" (id. at 46) for the samples designated "Dissolved." Presumably the Pace Analytical lab recognized this as an instruction to filter those samples; however, the uncertainty about filtration is compounded by Pace Analytical's "Sample Condition Upon Receipt" form (id. at 48), which has an ambiguous answer – "N/A" rather than "yes" or "no" – for the line which asks "Filtered volume received for Dissolved tests."

39. While it is possible that filtration was done as a routine procedure within EPA Method 903.1, §§ 8.1 and 8.4, such a procedure needs to be reconciled not only with the findings of Nelson et al. but also with the following observations about filtration in the Pennsylvania TENORM Report:

> Due to high solids content, the samples were not filtered in the field or at the laboratory. The aqueous portion was decanted from 10 of the 51 samples after they had been allowed to settle. The aqueous portion was analyzed for Ra-226 and Ra-228. These results are presented in Table 5-3 along with the original gamma spectroscopy results for the entire sample. The entire sample results include dissolved and undissolved Ra-226 and Ra-228 and are generally one to two orders of magnitude higher than analyses of only the aqueous phase, indicating that the Ra-226 and Ra-228 in these samples were mostly in the form of undissolved solids.
>
> (Pennsylvania TENORM Report at page 5-1.)

Judging from the above observations from the Pennsylvania TENORM Report, the filtration and/or decanting process performed on the Chemung samples was a crucial step, potentially involving a substantial loss of Radium-226, that should have been described in reasonable detail in the Chemung Leachate Test Report.

40. If Radium-226 activities in Chemung and Hakes leachate are indeed in the

neighborhood of 1000 or 6000 pCi/L, they would be much higher than any of the leachate results reported from Pennsylvania:

> Samples of leachate were collected from 51 landfills and analyzed using gamma spectroscopy for Ra-226 and Ra-228. The gamma spectroscopy results are presented in Table 5-1 for the 42 landfills not selected based on volume of O&G waste accepted and Table 5-2 for the nine landfills selected based on the volume O&G waste accepted. Radium was detected above the MDC value in 38 of 51 samples. Sample results from the 42 unselected landfills showed Ra-226 results that ranged from 36.5 to 416 pCi/L with an average of 116 pCi/L. Radium-226 results from the nine selected landfills ranged from 67.0 pCi/L to 378 pCi/L with an average of 125 pCi/L. Radium-228 results ranged from 2.50 to 55.0 pCi/L with an average of 11.9 pCi/L in the 42 unselected landfills. Radium-228 results from the nine selected landfills ranged from 3.00 pCi/L to 84.0 pCi/L with an average of 18.0 pCi/L.
>
> (Pennsylvania TENORM Report at page 5-1.)

41. The above comparison to Pennsylvania test results illustrates the potentially serious nature of errors in Hakes and Chemung sample collection, handling, and/or testing. However, the question of errors in the collection, handling, and/or testing of the Hakes and Chemung samples collected for Radium-226 analysis is entirely speculative in the absence of further testing. The above discussion of possible errors shows ways in which errors might have occurred but offers no clear evidence that errors actually occurred. The important point is that there are only two alternatives – or some combination of the two – that can explain the mismatch between the reported Radium-226 results and the reported Lead-214 and Bismuth-24 test results. Thus, if this alternative isn't true or predominant, the next one must be. As already noted, further testing could readily distinguish between these two alternatives.

Details of the only other alternative: High radon, relatively low radium, in sampled leachate

42. If Radium-226 *wasn't* actually present in the leachate in concentrations ranging up to ~1000 or ~6000 pCi/L, then the leachate must have contained high and apparently unrecognized levels of Radon-222 that were orders of magnitude higher than could be supported by secular

equilibrium with the reported levels of Radium-226. This possibility is plausible, given the high solubility of Radon-222 in water,[3] but further testing is needed to distinguish between this possibility and the possibility of underreported Radium-226. As noted above, further testing could be (or could have been) done in a straightforward manner by retesting existing samples.

43. In this alternative where Radon-222 activity substantially exceeds Radium-226 activity, the concentration and activity of Radon-222 must have been declining exponentially during the sample holding period of about 21 days in accordance with its 3.82-day half-life. During the 21-day holding period[4] – which corresponds to about 5.5 half-lives – the concentration and activity of Radon-222 must have declined by a factor of about 45. Calculating backward from a Radon-222 activity of ~1000 or ~6000 pCi/L at the time of testing (as indicated by the Lead-214 and Bismuth-214 results), the activity of dissolved Radon-222 in the leachate at the time of sample collection must have been ~45 times higher, or ~45,000 or ~270,000 pCi/L. If this alternative is true or predominant, the source and fate of such high levels of Radon-222 need to be investigated. The source is of course Radium-226, but where and how much? Radon-222 is a gas and thus able to migrate away from its parent Radium-226. The short half-life of Radon-222 requires that the parent Radium-226 must be nearby – in other words, within the landfill – so part of what's needed here is an analysis of the pathway within the landfill from the parent radium to its radon progeny. Also needed is a defensible inventory of the amount of Radium-226 within the

[3] For example, the online *Encyclopaedia Britannica* entry for Noble Gases lists the solubility of Radon-222 as about 230 cm^3 per liter of water at 20° C. This solubility limit can be combined with the specific activity of Radon-222 to express the solubility limit as an activity limit (>300,000 Ci/L) for Radon-222 in water at this temperature. Such a solubility limit, even if somewhat reduced as a result of other solutes present in landfill leachate, is orders of magnitude above the concentrations considered here and would therefore not be a limiting factor for Radon-222 solubility in the Chemung and Hakes leachate samples.

[4] Note that the sample holding time is unavailable for the 4Q 2014 Hakes leachate results because the relevant Hakes Leachate Test Report (Exhibit H) does not include the analytical data sheets that would show the Method 901.1 test dates. This is especially important for the reported 4Q 2014 activities of ~6000 pCi/L for Lead-214 and Bismuth-214. Presumably Pace Analytical and its customers understand the relevance of a 20- or 21-day ingrowth period and specified a sample holding time of least 20 or 21 days.

landfill that would produce enough Radon-222 to account for ~45,000 or ~270,000 pCi/L being present in the leachate at the time of sample collection.

44. Also needed is an analysis of the pathways by which Radon-222 and its progeny may leave the landfill, either escaping through landfill caps, etc., or dissolved in leachate taken offsite for treatment. Part of such analysis would assess the impacts of Radon-222 and its progeny to the environment and public health and safety. The activity of Radon-222 and its ability to flow as a gas would decline relatively quickly in accordance with its 3.82-day half-life, but the fate, transport, and impacts of its longer-lived progeny would need to be addressed.

45. The water-air partition coefficient for radon provides an approximate understanding of Radon-222 concentrations in the air/landfill gas mixtures that are in contact with Hakes and Chemung leachate. The partition coefficient, which can be calculated from the Weigel equation, depends not only on temperature but also on water salinity. See especially E.B. Lieberman, *Radon Solubility in Water as a Function of Salinity and Temperature*, M.S. Thesis, Florida State University (2013). While the coefficient's dependence on other constituents of the aqueous and gas phases has not been fully characterized, the water-air coefficient for radon can be used as an approximation.[5] At a temperature of 20º C, for example, the partition coefficient is about 0.2534. Thus, for air at equilibrium with water in which the dissolved Radon-222 activity is ~45,000 pCi/L (based on Chemung data) or ~270,000 pCi/L (based on Hakes data), the Radon-222 activity in air would be about 45,000/0.2534 = 177,000 pCi/L, or 270,000/0.2534 = 1.05 million pCi/L. Radon-222 concentrations such as 177,000 pCi/L or 1.05 million pCi/L are merely approximations of the levels that would be found in the air/landfill gas mixtures that are in contact with leachate, both within and outside the landfill – but these values provide an idea of the

[5] Alternatively, the Henry's Law constant for radon (0.0093 mol/L-atm at 25ºC) could be used for this purpose.

non-trivial Radon-222 concentrations that need to be assessed and addressed in the context of this high-radon/low-radium alternative. Within the landfill, one of the main pathways of interest is the aqueous and/or air pathway by which Radon-222 ingrowth from Radium-226 decay reaches the leachate. Other pathways of interest, both outside and inside the landfill, involve plume(s) of Radon-222 that offgas from the leachate.

46. There is currently no proof that this high-radon/low-radium alternative is the true or predominant alternative – but if it's not, then the previous one must be. As already noted, further testing could readily distinguish between these two alternatives.

47. It's important to recognize that the words "high-radon/low-radium" in this alternative refer only to the leachate. A relatively large amount of the parent radium must be present within the landfill even if this radium hasn't migrated into the leachate.[6] Solid materials such as radium and its chemical compounds would not tend to migrate if kept in a relatively dry section of a landfill, but gases such as radon may migrate through interconnected pore spaces within a landfill. Given the high density of radon relative to air or landfill gas, the radon produced from radium decay in a relatively dry upper section of a landfill may migrate downward toward – and into contact with – leachate. Some of the radon gas that comes into contact with leachate will then dissolve into the leachate, as indicated in the above discussion of the water-air partition coefficient for radon. Radon-222 concentrations in the air/landfill gas mixture above the leachate may potentially be as high as 177,000 pCi/L or 1.05 million pCi/L, as discussed above.

48. The radon migration process described above, coupled with radon's solubility in water and in water-based mixtures such as leachate, explains how leachate may become

[6] The short half-life of Radon-222 dissolved in the leachate requires that the parent radium must be nearby (i.e., within the landfill) in quantities sufficient to generate the high levels of Radon-222 present in the leachate at the time of sample collection (such as ~45,000 or ~270,000 pCi/L of dissolved Radon-222, as discussed above).

"high-radon/low-radium" while most of the parent radium remains in a relatively dry upper section of a landfill. In this manner, radon tends to pose a more immediate risk, but the radium itself will generally be a longer-term risk because of its long half-life. As a general rule, radiological risk from a given radionuclide will decline over time until it becomes negligible after several half-lives, e.g., ten half-lives. For Radium-226, with its 1600-year half-life, the relevant question of long-term risk is whether landfill integrity can be maintained for several half-lives, say 16,000 years.

49. Maintaining landfill integrity for thousands of years against erosion, animal burrows, inadvertent human intrusion, etc., appears unlikely, especially for a landfill not specifically designed for radionuclide disposal. While there are various methods of assessing the long-term risk from radionuclide disposal facilities, one of the crucial missing pieces of information at landfills such as Hakes and Chemung is an accurate inventory of radium contained within the landfill. As discussed above, leachate test results do not directly provide this information. At best, "high-radon/low-radium" leachate test results are proxy measures that show a need for additional testing. Such testing is needed to quantify the radium contained within a landfill that serves as the source of the radon measured in leachate.

Conclusions

50. High radionuclide concentrations in leachate are important not only as current measures of radiological contamination leaving the landfill, but also as proxy measures that may help quantify the source term or inventory of Radium-226 within the landfill. A good understanding of this inventory is needed to assess the landfill's potential long-term impacts and oversight needs. If gamma monitoring at the gate is not providing a reliable measure of Radium-226 entering the landfill, then proxy measures such as Radium-226 and/or Radon-222

concentration in leachate become increasingly important as screening-level indicators of the need for more direct sampling and testing of the contents of the landfill.

51. In assessing offsite radiological impacts to the environment and public health and safety, not only Radium-226 and/or Radon-222 but also the longer-lived progeny such as Lead-210 and Polonium-210 need to be considered.

52. The foregoing issues of high radionuclide concentrations in leachate need to be acknowledged and addressed in a transparent, deliberative, and defensible manner, preferably in the context of an EIS process.

53. Existing methods of monitoring trucks for Radium-226 at the Hakes and Chemung landfill gates are unreliable and unprotective. The so-called correlation between monitor readings and Radium-226 concentrations is flawed, such that waste truckloads containing identical Radium-226 concentrations may exhibit up to 60-fold variations in their monitor readings – and conversely, waste truckloads with up to 60-fold variations in their Radium-226 concentrations may exhibit the same or similar monitor readings. This problem stems from Lead-214 and Bismuth-214 gamma emissions, and from the fact that concentrations of these two radionuclides are not tied to Radium-226 concentrations but may vary widely from truckload to truckload. Using a radioisotope identification device at the landfill gate may help in addressing this problem and developing a reliable correlation. Such a correlation should not be pursued privately or confidentially; it needs to be developed in a transparent, deliberative, and defensible manner, preferably in the context of an EIS process.

54. This report is based on information available to me at this time. Should additional information become available, I reserve the right to determine the impact, if any, of the new information on my opinions and conclusions and to modify or supplement this report if necessary.

Sworn to before me this 18 day of January 2018.

Notary Public, State of New York

CARLYN J. SZAROWICZ
NOTARY PUBLIC STATE OF NEW YORK
ERIE COUNTY
LIC. #01SZ6320817
COMM. EXP. 3/9/2019

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Exhibit N - Chemung Leachate Radionuclide Analytical Results March 2015 through January 2017

Exhibit O - Answer posted 27 September 2005 by George Chabot, PhD, CHP, providing an expert online answer to the question, "What value should be taken as the average gamma energy of 226Ra?" at the Health Physics Society website [https://hps.org/publicinformation/ate/q4817.html]

Exhibit P - Uranium-238 decay series

Exhibit Q - Oak Ridge Institute for Science and Education (ORISE) presentation, Radiological and Chemical Properties of Uranium, available online from the U.S. Nuclear Regulatory Commission website (www.nrc.gov/docs/ML1122/ML11227A233.pdf)

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Exhibit W - Graph of time trends for Chemung leachate test results for Bismuth-214

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Exhibit Y - Graph of time trends for Chemung leachate test results for Radium-226 (tested by Method 901.1)

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